Dispersion in double-porosity unsaturated medium: from experiment towards modeling by homogenization

Tien Dung Tran Ngoc, Jolanta Lewandowska, Henri Bertin, Michel Vauclin, Lan Chi Do Hong

DISPERSION IN DOUBLE-POROSITY UNSATURATED MEDIUM: FROM EXPERIMENT TOWARDS MODELING BY HOMOGENIZATION

Tien Dung TRAN NGOC[1*], Jolanta LEWANDOWSKA[2], Henri BERTIN[3], Michel VAUCLIN[4] & Lan Chi DO HONG[5]

[1]*Bureau de Recherches Géologiques et Minières (BRGM) (French Geological Survey), Département Géothermie, 3 avenue Claude Guillemin, 45060 Orléans cedex 02, France. E-mail: td.tranngoc@brgm.fr*

[*]*Formely at 4.*

[2]*Laboratoire Sols, Solides, Structures-Risques (3S-R), UMR 5521 UJF, CNRS, INPG, 38041 Grenoble cedex 09, France. E-mail: jolanta.lewandowska@hmg.inpg.fr*

[3]*Laboratoire TRansferts, Ecoulements, FLuides, Energétique (TREFLE), UMR CNRS 8508, Université de Bordeaux, 33405 Talence cedex, France*

[4]*Laboratoire d'étude des Transferts en Hydrologie et Environnement (LTHE), UMR 5564 CNRS, INPG, IRD, UJF, 38041 Grenoble cedex 09, France.*

[5]*Vietnam National University of Hochiminh City (VNUHCM) Linh Trung, Thu Duc, Ho Chi Minh City, Viet Nam*

ABSTRACT

This study presents a macroscopic double-porosity dispersion model for unsaturated water flow conditions, developed by asymptotic homogenization method. The model shows a coupling between the concentrations in the micro- and macro-porosity domains (local non-equilibrium), resulting in an early breakthrough and a long tail effects. A series of one-dimensional dispersion experiments of passive tracer under unsaturated steady-state flow conditions in a physical model of double-porosity material, was performed. An excellent agreement between the numerical simulations and the experiments proves good predictive capacity of the developed model.

INTRODUCTION

Natural porous geomaterials are very often heterogeneous multiphase materials of complex structure. They are named "double-porosity" materials, when presenting a strong contrast in local hydrodynamic characteristics (aggregated soils, fractured porous rocks). In such cases, preferential solute transport in the macro-porosity domain with interactions/exchanges with the micro-porosity domain, are commonly observed. These interactions/exchanges are responsible for solute spreading in the local non equilibrium conditions and contribute to the non Fickian behaviour observed in the breakthrough curves.

Modeling unsaturated water flow and solute transport in such media remains an actual research subject, see for example [1]. In this paper we present a solute dispersion model for double-porosity media. The model is associated with the unsaturated water flow model [2] and was derived by using the up-scaling method (asymptotic homogenization technique). This method allows the derivation of a macroscopic model which is intrinsic to the material and independent of the macroscopic boundary conditions. No assumption about the mathematical form of the model is needed *a priori*. The obtained model was validated by a series of tracer transport experiments in a physical model revealing the double-porosity structure. This physical model was previously used in order to validate the transient unsaturated water flow model [2] for different scenarios of flow condition: infiltrations [3, 4] or drainage [5].

Tien Dung TRAN NGOC, Bureau de Recherches Géologiques et Minières (BRGM), Département Géothermie, 3 avenue Claude Guillemin, 45060 Orléans cedex 02, France.
et al.

DOUBLE-POROSITY PHYSICAL MODEL AND DISPERSION EXPERIMENTS

Double-Porosity Physical Model

A double-porosity physical model [3, 4, 5] was designed in order to mimic the behavior of the double-porosity geomaterials and enable to perform the dispersion experiments in fully controlled conditions. It was made of two porous materials: Hostun HN38 sand and solidified clayey spheres (Table 1). The double-porosity soil column (height L_c = 60 cm, inner diameter d = 6 cm) was obtained by arranging periodically the clayey spheres in the sandy matrix, layer by layer, followed by mechanical compaction. The medium has periodic structure (Figure 2). The contrast of the characteristic parameters of the two materials is the following:

- characteristic pore size ~1 : 200;
- water conductivity at saturation ~1 : 1500;
- effective diffusion coefficient at saturation ~1 : 10.

The selected physical properties are reported in Table I.

TABLE I. MEASURED PHYSICAL PROPERTIES OF THE MATERIALS

	ρ_s [g/cm^3]	d_{50} [μm]	n [-]	R [cm]	K_S [cm/s]
Sand HN38	2.65	162	0.400	-	2.87×10^{-3}
Clayey spheres	3.01	0.7*	0.348	3.2	1.96×10^{-6}

(ρ_s: skeleton specific density; d_{50}: mean grain diameter; *: mean pore size; n: porosity; R: radius of a sphere and K_S: conductivity at saturation.)

Dispersion Tests

An experimental set-up was designed in order to conduct a series of NaCl dispersion experiments in the unsaturated double-porosity column within the steady-state flow regime. Two principal stages of the experimental procedure concerned: i) the establishment of the permanent unsaturated flow, ii) the tracer transport by the fluid displacement. The water and the NaCl solution were introduced at the bottom of the column. While the unsaturated condition was controlled by measuring the water content, using the gamma ray attenuation technique, the evolution of NaCl concentration in the effluent was registered by the densimeter (Breakthrough Curve, BTC). Two different solicitation types (step-like and pulse-like) were used in the experiments. The detailed description of the experimental set-up and the procedure can be found in [6, 7]. The main characteristics of the experiments analyzed in this article are presented in Table II.

TABLE II. MAIN PARAMETERS OF EXPERIMENTS

	L [cm]	ϕ_1 [-]	ϕ_2 [-]	$\langle\theta\rangle$ [-]	$\langle v\rangle$ [cm/s]	Type	C_0 [g/l]
Test 1	51.0	0.490	0.510	0.348	3.44×10^{-4}	Step	5
Test 2	46.9	0.483	0.517	0.349	3.44×10^{-4}	Step	5
Test 3	46.9	0.483	0.517	0.349	3.44×10^{-4}	Pulse	5

(L: length of the column; ϕ_1 [-]: volumetric fraction of sand; ϕ_2 [-]: volumetric fraction of spheres; $\langle\theta\rangle$: mean water content of the double-porosity medium; $\langle v\rangle$: imposed Darcy water flow velocity; C_0: concentration of NaCl applied at the inlet to the column.)

Qualitative Analysis of The Dispersion Tests

Figure 1 shows the comparison between the behavior of sand (which is considered as simple-porosity material), and sand (of the same porosity), containing porous spheres (double-porosity material) [6]. The quantitative analysis of the two breakthrough curves gives three principal conclusions concerning the behavior of the double-porosity medium:

- The breakthrough curve is asymmetric.
- The acceleration of the appearance of concentration (early breakthrough) is observed.
- The concentration approaches the imposed value at a long time (tailing).

All these features are characteristic for the non Fickian behavior which can not be described by classical dispersion models.

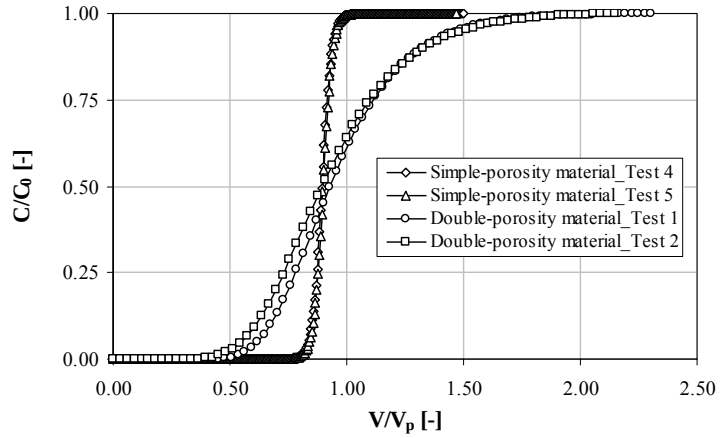

Figure 1. BTCs observed in dispersion tests with the simple- porosity (sand, Test 4 & 5) and double-porosity material (sand with porous spheres, Test 1 & 2).

MODELING BY ASYMPTOTIC HOMOGENIZATION

The development of the macroscopic model of solute transport in unsaturated double-porosity media obtained by the application of the homogenization technique [8, 9, 10], will be presented briefly. The detailed analysis can be found in [7].

General Assumptions and Estimations

The general assumptions are:

- the medium is rigid and its structure is periodic,
- there exists a scale separation, defined by the small parameter $\varepsilon = \frac{\ell}{L} << 1$

where ℓ [L] and L [L] are the dimensions of the period and macroscopic domain (column length), respectively,

- the more conductive sub-domain is connected,
- the contrast of the hydraulic conductivity in the macro- and micro-porosity domains is $\frac{K_2}{K_1} = O(\varepsilon^2)$.

The time of observation of the phenomenon is of the order of $O(10^5$ s) (see Figure 3) which corresponds to the order of the time of transport by convection in the macro-porosity domain at the scale L:

$$t_c = \frac{L}{v_{1c}} = \frac{51 \times 0.355 \times 0.49}{3.44 \times 10^{-4}} = 25789\ s\,. \tag{1}$$

Two additional assumptions concern the Péclet number of the transport through the macro-porosity domain and the ratio of the local diffusion/dispersion coefficients of the micro- and macro-porosity domains:

$$Pe_1 = \frac{\ell\, v_{1c}}{D_{1c}} = O(1) \text{ and } \frac{D_{2c}}{D_{1c}} = O(\varepsilon)\,. \tag{2}$$

Using the above assumptions and estimations, the homogenization problem can be formulated for the non-dimensional variables in the form:

$$\varepsilon \frac{\partial(\theta_1^*\, C_1^*)}{\partial t^*} = \nabla \cdot \left(\boldsymbol{D}_1^*(\theta_1)\, \nabla C_1^* - \boldsymbol{v}_1^*\, C_1^* \right) \text{ in } \Omega_1, \tag{3}$$

$$\frac{\partial(\theta_2^*\, C_2^*)}{\partial t^*} = \nabla \cdot \left(\boldsymbol{D}_2^*(\theta_2)\, \nabla C_2^* \right) \text{ in } \Omega_2, \tag{4}$$

$$\left(\boldsymbol{D}_1^*(\theta_1)\, \nabla C_1^* - \boldsymbol{v}_1^*\, C_1^* \right) \cdot \boldsymbol{N} = \varepsilon \left(\boldsymbol{D}_2^*(\theta_2)\, \nabla C_1^* \right) \cdot \boldsymbol{N} \text{ on } \Gamma, \tag{5}$$

$$C_1^* = C_2^* \text{ on } \Gamma, \tag{6}$$

where "*" is denoted for the dimensionless quantities; C_1 and C_2 are the concentration in Ω_1 and Ω_2 respectively; $\boldsymbol{D}_1$ is the local dispersion tensor in Ω_1; $\boldsymbol{D}_2$ is the diffusion tensor in Ω_2 ; $\boldsymbol{v}_1$ is the velocity in the macro-porosity domain; θ_1 and θ_2 are the water contents in Ω_1 and Ω_2 respectively; Ω_1 and Ω_2 are the macro- and micro-porosity domains in the period Ω (Figure 2); $\boldsymbol{N}$ is the unit vector normal to the interface Γ; and the t is the time variable.

Macroscopic Model of Dispersion in A Double-Porosity Medium

The macroscopic dispersion model obtained by homogenization is composed of two coupled equations:

$$\phi_M \frac{\partial(\theta_M\, C_M)}{\partial t} = \nabla \cdot \left(\boldsymbol{D}_{\text{disp}}(\theta_M)\, \nabla C_M - \langle \boldsymbol{v} \rangle\, C_M \right) - \frac{\partial \langle \theta_m C_m \rangle}{\partial t}\,, \tag{7}$$

$$\frac{\partial(\theta_m C_m)}{\partial t} = \nabla \cdot \left(\boldsymbol{D}_m(\theta_m)\, \nabla C_m \right), \tag{8}$$

$$C_M = C_m \text{ on the interface,} \tag{9}$$

where C_M and C_m [ML^{-3}] are the concentration in the macro- and micro-porosity respectively; θ_M and θ_m [-] are the water content in the macro- and micro-porosity

respectively; ϕ_M [-] is the volumetric fraction of the macro-porosity domain; $< \boldsymbol{v} >$ [LT^{-1}] is the Darcy velocity; $\boldsymbol{D}_{disp}$ [L^2T^{-1}] is the dispersion tensor of the double-porosity medium; $\boldsymbol{D}_m$ [L^2T^{-1}] is the effective diffusion tensor in the micro-porosity; and t [T] is the time variable.

The model (7)-(9) reveals the local non-equilibrium due to the presence of two concentration fields. The effective parameter of the model is the dispersion tensor that depends on the local dispersion tensor and the microstructure of the double-porosity medium. It can be calculated from the solution of the local boundary value problem [7].

NUMERICAL SIMULATIONS

Numerical Implementation

The double-porosity dispersion model (7)-(9) was implemented using the commercial code COMSOL Multiphysics. This code enables coupling between the two processes (dispersion and diffusion) taking place in two different domains: the macro-porosity and micro-porosity domains, respectively. It was done by defining the coupling variables: the concentration and the flux at the interface. The details concerning the implementation can be found in [7].

Comparison Between Numerical Simulations & Experimental Observations

In order to validate the developed theoretical model we carried out the numerical simulations of the experiments and we compared the results with the observations. This process consists of two stages: calibration and validation.

We calibrated the dispersion coefficient in the double-porosity medium, using the experimental result of Test 1. Figure 3 presents the normalized concentration evolution of the simulation and experiment with time, at the outlet of column for Test 1. The dispersion coefficient in the double-porosity medium was obtained, $D_{disp} = 2.347\times10^{-4}$ cm^2/s. Note that the diffusion coefficient in the solidified clay was determined from the literature (for bricks), $D_2 = 7.345\times10^{-7}$ cm^2/s.

The obtained parameters of the model were used to simulate two other independent experiments: Test 2 (step-like) and Test 3 (pulse-like). Figure 4 and 5 show an excellent agreement between the simulated and experimental BTCs. Thus, it can be concluded that the model was successfully validated. Moreover, the determined dispersion coefficient can be considered as an intrinsic parameter, corresponding to a given flow velocity and a given water content.

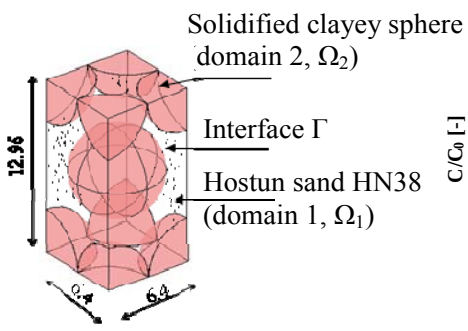

Figure 2. Geometry of the period (Ω) (unit in mm).

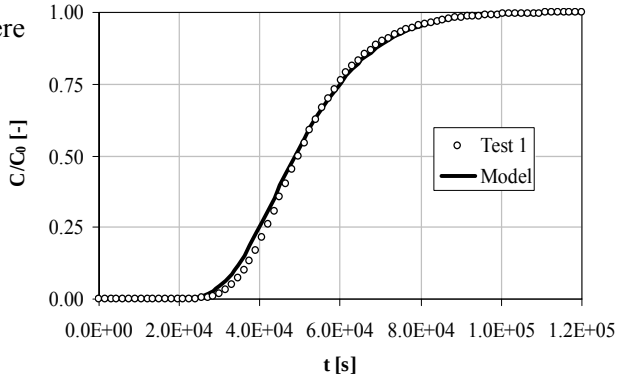

Figure 3. The BTCs in the calibration stage of the double-porosity model.

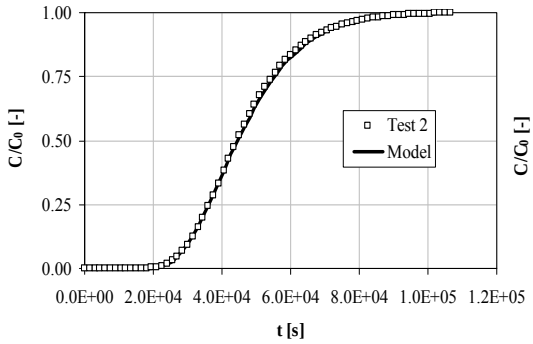

Figure 4. Model (line) and experiment (symbols) BTCs in the step-like test.

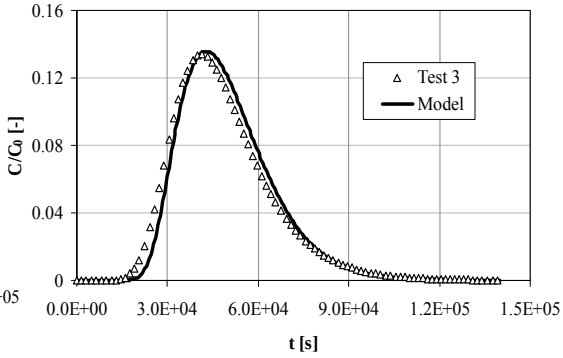

Figure 5. Model (line) and experiment (symbols) BTCs in the pulse-like test.

CONCLUSION

The double-porosity dispersion model obtained by the asymptotic homogenization method proved to be able to describe the solute transport in the double-porosity medium. It was evaluated against a series of laboratory dispersion experiments on a physical model of double-porosity. The numerical simulations showed an excellent agreement with the experimental data. The complete analysis will be presented in the forthcoming journal paper.

Acknowledgement: We acknowledge the financial support from the GDR "Hydrodynamique et Transferts dans les Hydrosystèmes Souterrains" and the BDI-PED doctoral fellowship from the CNRS, France granted to the first author.

REFERENCES

1. Köhne, J. M., S. Köhne, and J. Šimůnek. 2009. "A review of model applications for structured soils: A) Water flow and tracer transport," *J. Contam. Hydro. 104: 4-35.* doi: 10.1016/j.jconhyd.2008.10.002
2. Lewandowska, J., A. Szymkiewicz, K. Burzynski, and M. Vauclin. 2004. "Modeling of unsaturated water flow in double - porosity soils by the homogenization approach," *Adv. in Water Resour*. , 27:283-296.
3. Lewandowska, J., A. Szymkiewicz, W. Gorczewska, and M. Vauclin. 2005. "Infiltration in a double-porosity medium: Experiments and comparison with a theoretical model," *Water Resour. Res.,* 41(2):1-14.
4. Szymkiewicz, A., J. Lewandowska, R. Angulo-Jaramillo, J. Butlanska. 2008. "Two-scale modelling of unsaturated water flow in double-porosity medium under axisymmetric conditions," *Canadian Geotechnical J.,* 45: 238-251.
5. Lewandowska, J., T. D. Tran Ngoc, M. Vauclin, and H. Bertin. 2008. "Water drainage in double-porosity soils: experiments and micro-macro modeling," *J. Geotech. & Geoenvir. Eng.,* 134(2): 232-243.
6. Tran Ngoc, T. D., J. Lewandowska, and H. Bertin. 2007. "Etude expérimentale de la dispersion dans un milieu à double-porosité," *Congrès Français des Mécaniques,* Grenoble, CD-Rom.
7. Tran Ngoc, T. D. 2008. "Transport de solutés dans un milieu à double-porosité non saturé. Modélisation par homogénéisation et application," *Ph.D thesis,* Université Joseph Fourier-Grenoble, 185 p (in French).
8. Bensoussan, A., J.L. Lions, and G. Papanicolaou. 1978. "Asymptotic analysis for periodic structures,". North-Holland.
9. Sanchez-Palencia, E. 1980. "Non-homogeneous media and vibration theory," Vol. 127 of *Lecture Notes in Physics,* Springer-Verlag, Berlin.
10. Auriault, J.L. 1999. "Heterogeneous medium. Is an equivalent macroscopic description possible?," *Int. J. Engng Sci.,* 29: 785-795.